

Preliminary self-sealing test results for selected argillaceous media in England

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Abstract

There is international consensus that geological disposal is the safest permanent solution to manage the most hazardous radioactive waste. The UK is considering argillaceous rocks of the Mercia Mudstone Group and the Ancholme Group as potential host rock for geological disposal. A preliminary programme of laboratory testing aiming at investigating the self-sealing mechanisms of argillaceous media in England was carried out at the Rock Mechanics, Engineering Geology and Geotechnical laboratory at the University of Leeds. Mercia Mudstone Group rocks of Triassic age from sites across England such as Sidmouth, Newark-on-Trent, and Teesside as well as Ancholme Group rocks of Jurassic age from Cambridgeshire have been considered and tested in this study. In addition to self-sealing experiments, basic rock characterisation testing such as Quantitative X-Ray Diffraction, water content, durability, and strength tests have been carried out. The results indicate the Mercia Mudstone Group rocks have a low durability and are predominately moderately weak while the Ancholme Group rocks have a very low durability and are very weak. Both the Mercia Mudstone and Ancholme Group rocks show self-sealing behaviour but differ according to their composition, specifically the type and quantity of cementing. From the perspective of geological disposal of radioactive waste, the preliminary results presented demonstrate that a range of argillaceous media in England can self-seal.

Keywords

Self-sealing, Mercia Mudstone Group, Ancholme Group, radioactive waste disposal, Geological Disposal Facility.

1 Introduction

The United Kingdom (UK) has been generating radioactive waste from power generation, industry, medicine, and defence for over 60 years, (GOV.UK, 2022). There is international consensus that geological disposal is the safest permanent solution to manage the most hazardous radioactive waste, (GOV.UK, 2018). Currently (2024) the UK is considering argillaceous rocks of the Mercia Mudstone Group (MMG) and the Ancholme Group (AmG) as potential host rocks for a Geological Disposal Facility.

One key factor in determining the suitability of argillaceous media as a potential host rock for geological disposal of radioactive waste is their ability to self-seal, along with the very low material permeability, fluid transport predominately by diffusion, strong retention capacity, and continuity of deposits, (Norris, 2019).

Self-sealing was defined by Tsang and Bernier (2005) as “the reduction of fracture permeability (transmissivity) by any hydromechanical, hydrochemical, or hydrobiochemical processes”. Self-sealing in argillaceous media is an important mechanism for many engineering applications including disposal of radioactive waste, (NEA, 2010 and Zhang and Talandier 2023), mining (Nagra, 2020), and hydrocarbon seals, (Fisher et al. 2021).

Significant work has been carried out globally to investigate self-sealing of argillaceous media as such, self-sealing mechanisms are a well-understood phenomenon in certain geological environments, (NEA, 2010). Although the fundamental questions about ‘if’ and ‘how’ self-sealing occurs have been substantially answered there are clear gaps in knowledge around the rate and magnitude of self-sealing of the MMG and AmG. The objective of our research is to measure the magnitude and rapidity of self-sealing in selected argillaceous media which are being considered as potential host rocks for a GDF in England and Wales.

2 Experimental work

Experimental work aiming at investigating self-sealing mechanisms of UK argillaceous materials was conducted and it involved both field work and laboratory testing carried out at the Rock Mechanics, Engineering Geology and Geotechnical (RMEGG) laboratory at the University of Leeds.

2.1 Fieldwork and geological materials

The following materials have been considered as part of this study:

1. The MMG is Triassic in age (approximately 247 Ma to 201 Ma) and was deposited in a series of fault bounded basins in the interior of Pangea, north of the Variscan mountain belt, Hobbs (2002) and Howard (2008). At the time the United Kingdom was at a similar geographical location to northern Africa (20 to 30 degrees north) with a monsoonal climate resulting in high intensity precipitation events followed by extended periods of aridity, Hobbs (2002) and Jeans (2006). Fluvial deposition of the underlying Sherwood Sandstone Group gave way to subaqueous hypersaline and evaporitic mudflat environments, leading to four main depositional processes, Howard (2008); settling of clay and silt size particles in saline water bodies, sheet wash deposition of silt and sand during flash floods, aeolian deposition of predominately fine sand, silt, and clay size particles on wet mudflats, and precipitation of evaporites such as halite and gypsum from water bodies and groundwater.
2. The AmG is Jurassic in age (approximately 166 Ma to 152 Ma), and developed between Norfolk and Humberside, Woods (2022). The AmG was deposited in a marine environment within the East Midlands Shelf when the United Kingdom was in a similar geographical location to the Mediterranean Sea (30 to 40 degrees north) with a warm and humid climate, Barron (2012).

Samples of MMG material are composed of predominately siltstone and claystone and were collected from various locations across the UK. Block samples were taken from Jacobs Ladder Beach, west of Sidmouth (Sid Mudstone Member of the Sidmouth Mudstone Formation), block samples from Bantycok Mine, southeast of Newark-Upon-Trent (Branscombe Mudstone Formation, Blue Rock Muck), and core samples taken from Redcar, Teesside (undifferentiated), presented in Fig. 1.

Samples of AmG material are composed of shelly, calcareous claystone and claystone. These samples were collected from the following locations; core samples were taken from the Fens Reservoir site north of Chatteris (undivided Oxford Clay Formation), block samples from Must Farm Quarry, southeast of Peterborough (shelly calcareous Peterborough Member of the Oxford Clay Formation), and core samples from the Lincs Reservoir site east of Grantham, (undivided Oxford Clay Formation – not reported on in this paper), shown in Fig. 1.

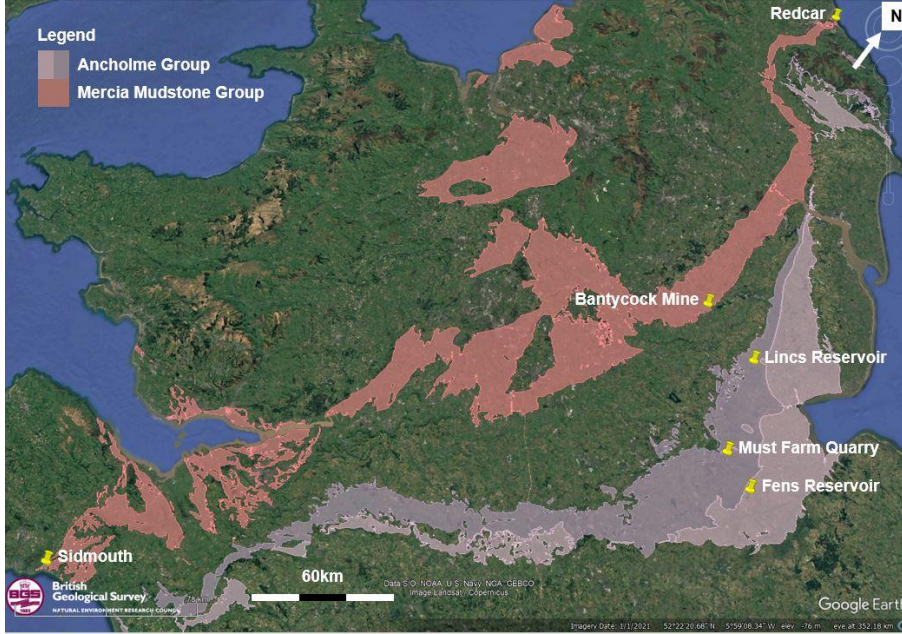


Fig. 1: Location of sampling sites. The British Geological Survey onshore geology 1:625,000 scale DiGMapGB625 layer for TRIA-MDSS (Triassic Rocks undifferentiated), KLOX-MDSS (Kellaway and Oxford Clay formations), WWAK-MDSS (West Walton, Amphill Clay, and Kimmeridge Clay formations). MDSS = mudstone, siltstone, and sandstone. © Google Earth, 'Contains British Geological Survey materials © UKRI 2024'.

2.2 Methods for laboratory testing

Specimen preparation required careful consideration due to the sensitive nature of the material and their vulnerability to environmental change. After significant trials it was found that MMG specimens could be satisfactorily prepared using dry coring techniques with a pillar drill. Whereas the more sensitive AmG materials required hand trimming using saws and knives and finishing on a lathe.

Experiments were carried out to determine the moisture content, slake durability, and strength (point load and Uniaxial Compressive Strength, UCS) in accordance with the relevant ISRM suggested methods. Quantitative X-Ray Diffraction (QXRD) determinations were carried using methods set out in Hillier (2000). The preliminary results are presented in Section 3.

Self-sealing tests were conducted on 50 mm diameter, approximately 50 mm long cylindrical specimens. To create the 'discontinuity' plane, the specimens were sawed longitudinally down the centre of the core using a lapidary saw. Before installing the sample in the triaxial cell, the discontinuity was propped open with 1mm thick spacers and glued back together. The self-sealing test was carried out in a Wykeham Farrance Tritex 50 soil triaxial rig using 200 kPa cell. The experiment comprised passing tap water through the propped discontinuity under a small hydraulic pressure equivalent to 1 m head of water to minimise the potential of erosion, shown in Fig. 2. Self-sealing was measured by recording the time taken for a volume of water to flow through the discontinuity. The fracture hydraulic conductivity was calculated using the equations derived from Darcy, see Eq. 1. The results are presented in Fig. 4 in Section 3.

$$K_f = \frac{-Q_f}{2b \cdot w \cdot \frac{dh}{dl}} \quad (1)$$

Where K_f Hydraulic conductivity (ms^{-1})
 Q_f Flow through a fracture (m^3s^{-1})
 $2b$ Discontinuity aperture (m)

w Width of discontinuity (m)
 dh/dl The dimensionless hydraulic gradient in the direction of decreasing head

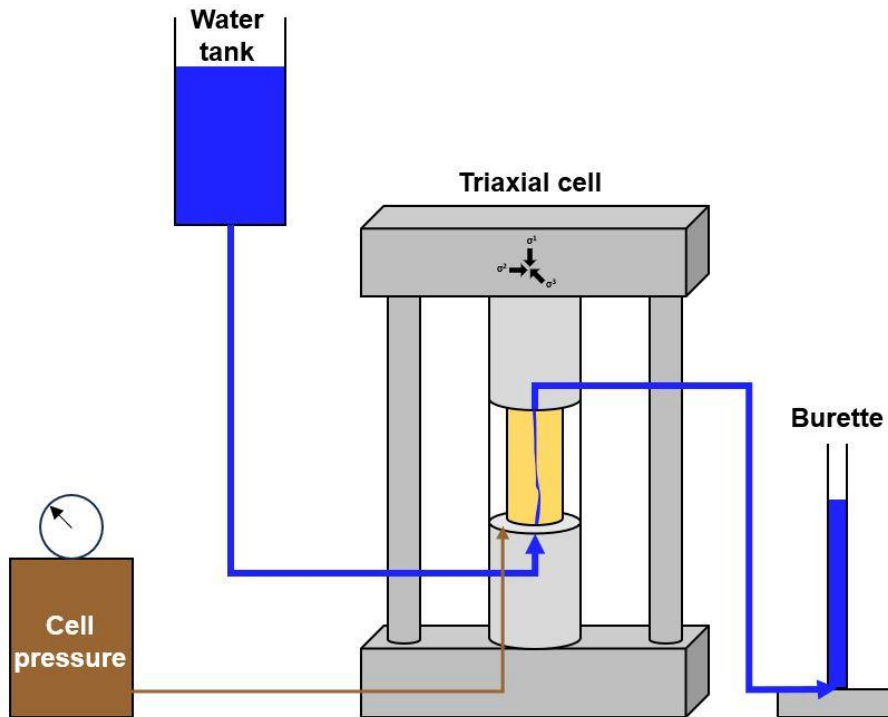


Fig. 2: Schematic of self-sealing experiment apparatus used at RMEEG laboratory facilities at the University of Leeds.

3 Laboratory results

The preliminary results of selected testing carried out at the RMEEG laboratory at the University of Leeds to investigate self-sealing of MMG and AmG rocks are presented herein.

3.1 Mercia Mudstone Group

The water content test results in Table 1 indicate the MMG samples recovered from surface exposures had a significantly lower moisture content (2.53 % to 5.75 %) than the MMG samples recovered from core (11.86 %).

The slake durability tests carried out on samples of MMG rocks indicate materials of very low to low durability (39 % to 58 %) shown in Table 1.

Point load tests carried out on specimens of MMG rocks indicate materials with an Unconfined Compressive Strength (UCS) of between 6.9 and 31.8 MPa, indicating weak to moderately strong materials, see Table 1.

Table 1: Summary of classification and strength test results – Range, (average if applicable), [number of tests].

Location (Stratigraphic unit)	Water content (%)	Slake durability (Id2, %)	UCS (MPa)
Sidmouth (Sid Mudstone Mb., Sidmouth Fm., MMG)	5.40-6.11, (5.75), [2]	55.5, [1]	13.2, [1] [†]
Bantock Mine (Branscombe Mudstone Fm., MMG)	2.31-2.74, (2.53), [2]	12.6-39.4, (26.0), [2]	16.1-26.4, (21.3), [5] [†]
Redcar (Undivided Mercia Mudstone, MMG)	7.39-15.26, (11.86) [3]	58.4, [1]	6.9-31.8, (19.0), [9] ^{†‡}
Fens Reservoir (undivided Oxford Clay, AmG)	16.11-16.97, (16.5) [2]	15.13-16.26, (15.70), [2]	1.1-1.7, (1.4), [2]
Must Farm Quarry (Peterborough Mb, Oxford Clay Fm., AmG)	11.99-17.34, (13.43) [8]	55.6, [1]	3.9, [1]

[†]Calculated from axial PLT using a multiplier of 16. [‡]Calculated from irregular lump PLT using a multiplier of 16.

Quantitative X-Ray Diffraction tests were carried out (Fig. 3). The results were broadly consistent with published literature, (Hobbs et al, 2002). The following three key observations from the XRD analysis can be made:

1. MMG specimens are cemented with dolomite.
2. The results show the dominant potential swelling clay mineral (Smectite) is bound with Illite (a non-swelling mineral) indicating a potential for swelling.
3. Haematite is the dominant iron mineral in specimens of MMG.

Illite-smectite was not identified in either sample from Sidmouth and it is unlikely that this is representative of the materials. It is assumed; however, that these clay minerals were reported as mica. This assumption is considered reasonable as the results indicate elevated quantities of mica.

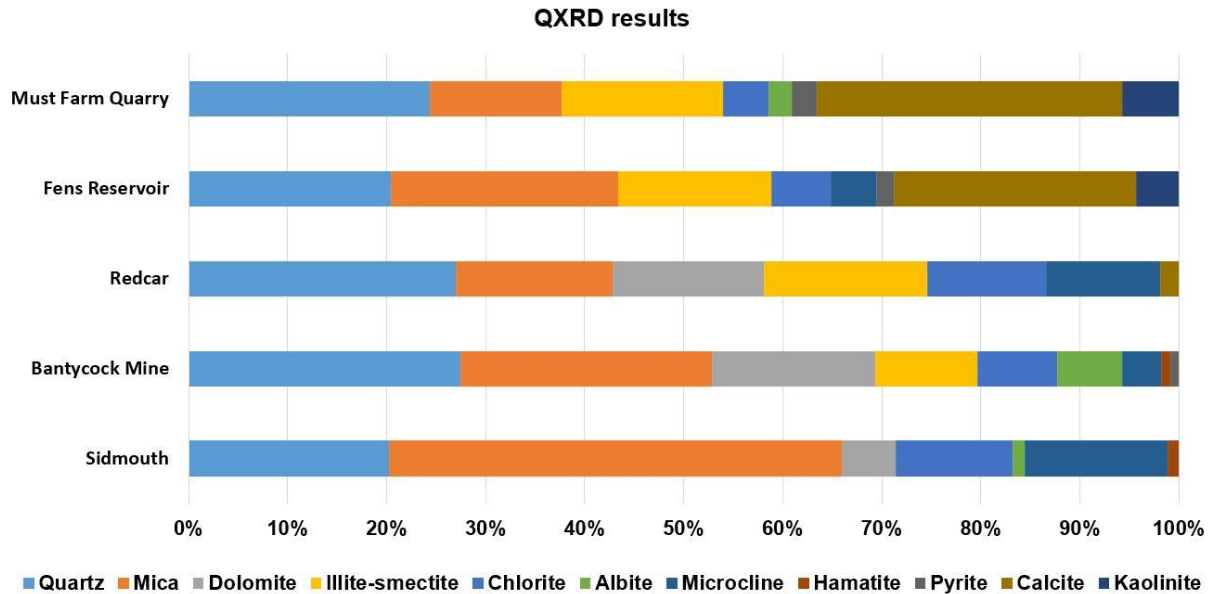


Fig. 3: A summary of QXRD results (averaged for each location).

The results of the flow tests carried out on specimens of MMG materials indicate that self-sealing is occurring in three broad stages, see Fig. 4. An early stage (approximately 0-24 hours), where initially there is little or no response to the water, a middle stage (approximately 24-96 hours) where there is a rapid reduction in fracture hydraulic conductivity, and a late stage (approximately >96 hours) when reduction in hydraulic conductivity stabilises at a reduced rate. The test results show a sudden reduction in fracture hydraulic conductivity that we have described as ‘blooming’.

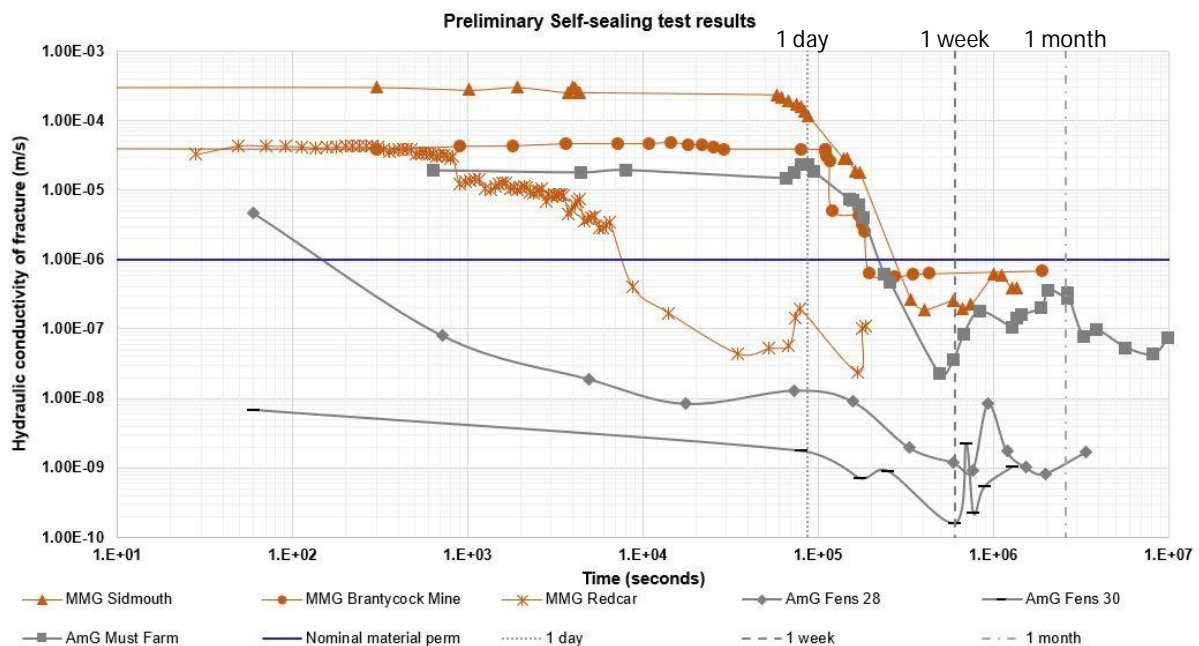


Fig. 4: Self-sealing test results.

3.2 Ancholme Group

The water content test results for samples of AmG are similar for both the specimens taken from core and surface exposures (16.5 % and 17.9 %), see Table 1. The sample recovered from the quarry was taken within a few hours of excavation and wrapped with successive layers of clingfilm and tin foil therefore reducing the exposure of the material to drying out. In addition, specimen tested was taken from approximately the centre of the block further reducing the impact of exposure to the atmosphere and drying.

The slake durability tests carried out on samples of AmG rocks indicate materials of low durability from the Must Farm location (55%) and very low durability from the Fens Reservoir site (15.13 % and 16.26 %), see Table 1. The higher durability of the sample from Must Farm Quarry is likely to be due to the presence of shelly material and greater carbonate cementing than in the samples from the Fens Reservoir site.

UCS tests carried out on specimens of AmG rocks indicate materials with a strength of between 1.1 MPa and 1.7 MPa (Fens Reservoir site) and 3.9 MPa for shelly carbonate specimens (Must Farm Quarry), indicating very weak materials.

Quantitative X-Ray Diffraction tests were carried out, see Fig. 3. They were broadly consistent with published literature, (Jeans and Merriman, 2006). Three key observations from data are:

1. AmG specimens were cemented with calcite.
2. The results show the dominant potential swelling clay mineral (Smectite) is bound with Illite (a non-swelling mineral) indicating a potential for swelling.
3. Pyrite is the dominate iron mineral in specimens of AmG.

The results of the flow tests carried out on specimens of AmG materials indicate that self-sealing is a more constant process that initiates almost immediately after the specimen is placed in the triaxial cell, see Fig. 4. It is interesting to note that the time required to reduce the hydraulic conductivity to the lower stabilised rate is similar to that of the MMG specimens at around 96 hours after the start of the test.

3.3 Discussion

The results of the laboratory experimentation indicate that specimens tested from block samples were drier than from the core. The reduced water content results from block samples indicate that the samples have dried out due to exposure to the atmosphere, whereas the core samples were preserved by wrapping in successive layers of cling film and tin foil immediately after logging, reducing the impact of exposure to air.

The slake durability tests indicate that specimens composed of argillaceous material with dolomite cementing i.e., MMG, or shelly calcareous material i.e., from Must Farm Quarry, are more durable than the specimens tested from the Fens Reservoir site that are predominately composed of claystone with lower levels of calcareous cementing. As with the durability testing, the strength tests indicate that the finer-grained and less calcareous AmG material is weaker than coarser-grained dolomite cemented MMG, and the shelly calcareous AmG.

The results of the flow tests carried out on specimens of MMG materials and the shelly, carbonate-rich specimen from Must Farm Quarry all show a sudden reduction in fracture hydraulic conductivity at around 24 hours after the start of the test that we have described as 'blooming'. The 'blooming' stops around 96 hours after the start of the test. It is thought that the 'blooming' effect could be a function of either, a delayed response due to the time required for the clay mineral swelling pressures to overcome the tensile strength of the grain boundaries or a time dependent response due to the time required to initiate clay mineral swelling, (Auvray, 2015 and Giot 2019). The results of the flow tests carried out on specimens from the Fens Reservoir site indicate that self-sealing is a more constant process that initiates whilst the test is being set up. The time required to reduce the hydraulic conductivity to the lower stabilised rate is similar in both the MMG and AmG specimens.

The results show distinct geomechanical and hydrogeological behavioural differences between specimens of MMG argillites and AmG claystones. This is not unexpected as the materials are

lithologically different and have different geological and stress histories. In summary, the MMG Rocks are composed of a higher proportion of coarse material such as silt and sand-sized particles that have been cemented by dolomite whereas the AmG rocks are finer grained and are cemented by calcite. In addition, the post-deposition depth of burial of the MMG was estimated to be between 2,000 m and 2,500 m and was heated to around 80 °C, Armitage et al. (2016), Holford et al. (2009), and Jackson and Mulholland (1993), whereas the AmG were buried to around 1,700 m, and were heated to around 60 °C, Green (2018). The geological stress histories have influenced the composition and hydro-geomechanical properties of the material.

The analysis presented, although, useful has several limitations, not least its preliminary status representing early findings from an ongoing research project. Other limitations identified include some elements of the sampling location and extraction techniques, as well as the sampling and preservation protocols, which were outside of the influence of the researchers so may not have been carried out to optimise a laboratory testing programme. Natural rocks are not uniform sometimes subtle variations mean that the specimens were not homogeneous. Therefore, careful consideration is required when comparing results from different experiments at the same site as well as comparing the results from the same experiments at different sites.

4 Conclusions

Preliminary information from a programme of laboratory experiments investigating self-sealing of Mercia Mudstone Group and Ancholme Group rocks is presented. The results indicate that minor variations in grain size, cementing, and cementing material have a significant impact on geomechanical and hydrogeological behaviour of material regardless of age or sampling location.

From the perspective of geological disposal of radioactive waste, the preliminary results presented demonstrate that a wide range of argillaceous media in England and Wales can self-seal.

The research is at an early stage, further work planned include increasing the number of tests carried out using the experiments described, broadening the range of experiments to include swelling tests, material porosity and permeability, and strength testing. In addition, refinements to the experimental processes will be carried out i.e., using clay mounts and glycolisation to ensure a more precise QXRD analysis.

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